

# New constraints on the delivery of cometary water and nitrogen to Earth from the $^{15}\text{N}/^{14}\text{N}$ isotopic ratio

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The delivery of cometary water and nitrogen to Earth

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## ABSTRACT

New independent constraints on the amount of water delivered to Earth by comets are derived using the  $^{15}\text{N}/^{14}\text{N}$  isotopic ratio, measured to be roughly twice as high in cometary CN and HCN as in the present Earth. Under reasonable assumptions, we find that no more than a few percent of Earth's water can be attributed to comets, in agreement with the constraints derived from D/H. Our results also suggest that a significant part of Earth's atmospheric nitrogen might come from comets. Since the  $^{15}\text{N}/^{14}\text{N}$  isotopic ratio is not different in Oort-cloud and Kuiper-belt comets, our estimates apply to the contribution of both types of objects.

*Keywords:* COMETS ; EARTH ; ORIGIN, SOLAR SYSTEM

## 1 Introduction

The origin of water on Earth is still puzzling (e.g. the reviews by Owen and Bar-Nun, 1998; Robert, 2001; Drake, 2005; Javoy, 2005; Marty and Yokochi, 2006, and references therein). At one end of the models, the high temperature in the inner accretion disk hampered hydrous phases to exist so that external sources were needed, the so-called late veneer of comets, asteroids and/or meteorites suggested by lunar cratering (e.g. Gomes et al., 2005). At the other end, Earth accreted hydrous silicate phases, or grains having adsorbed water from the solar nebula. The D/H isotopic ratio plays a key role in constraining the models. The comparable D/H ratio of Earth's oceans and carbonaceous chondrites (the only ones to contain enough water) points towards a meteoritic veneer, while the cometary D/H ratio, twice as high as the terrestrial one, would require a mixture of cometary water with a roughly equal amount of primitive indigenous D-poor terrestrial water. While the abundance of noble gases in the Earth's atmosphere suggests that only small amounts of cometary volatiles were brought to Earth, the  $^{187}\text{Os}/^{188}\text{Os}$  ratio apparently rules out the carbonaceous chondrites as the main component of the veneer (Drake, 2005, and references therein). However, most of these arguments have shortcomings (see discussions by Drake, 2005; Javoy, 2005; Marty and Yokochi, 2006; Dauphas, 2003; Genda and Ikoma, 2008). In particular, D/H has only been measured in Oort-Cloud comets and it is not excluded that other types of comets –or other reservoirs of hydrogen in comets– exhibit different D/H ratios. Additional constraints are therefore mandatory.

Recently, we found that the isotopic ratio  $^{15}\text{N}/^{14}\text{N}$  is consistently twice as high in cometary CN and HCN as in the Earth's atmosphere and surface (Arpigny et al., 2003; Bockelée-Morvan et al., 2008; Manfroid et al., 2009). Since any delivery of water by comets would necessarily be accompanied by a delivery of nitrogen, the observed difference between terrestrial and cometary  $^{15}\text{N}/^{14}\text{N}$  can induce a measurable isotopic shift and provide independent constraints on the amount of water and nitrogen delivered by comets to Earth's oceans and atmosphere, as detailed below. The interest of considering the nitrogen isotopic ratio has been recently emphasized by Marty and Yokochi (2006).

## 2 Constraints from D/H : framework and previous results

We assume that cometary  $\text{H}_2\text{O}$  and  $\text{HDO}$  add to a primitive mixture on Earth to give the amounts presently measured in Earth's water. We have then

$$\begin{aligned} n_p(\text{H}_2\text{O}) + n_c(\text{H}_2\text{O}) &= n_t(\text{H}_2\text{O}) \\ n_p(\text{HDO}) + n_c(\text{HDO}) &= n_t(\text{HDO}) \end{aligned} \quad (1)$$

where  $n$  is the abundance in number, the suffix  $p$ ,  $c$ , and  $t$  respectively referring to primitive Earth, cometary and terrestrial (present Earth) values. From these equations we derive

$$\frac{n_c(\text{H}_2\text{O})}{n_t(\text{H}_2\text{O})} = \frac{(\text{D}/\text{H})_p - (\text{D}/\text{H})_t}{(\text{D}/\text{H})_p - (\text{D}/\text{H})_c} \quad (2)$$

which provides the proportion of Earth's water due to comets as a function of the isotopic ratio D/H in water.

The present Earth D/H ratio is fairly well known and measured to be  $(\text{D}/\text{H})_t = 1.49 \pm 0.03 \cdot 10^{-4}$  for water stored at the Earth's surface, mainly in the oceans (Lécuyer et al., 1998). For cometary water we have  $(\text{D}/\text{H})_c = 3.1 \pm 0.3 \cdot 10^{-4}$  from in situ measurements of comet 1P/Halley (Balsiger et al., 1995; Eberhardt et al., 1995) and from remote observations of three Oort-Cloud comets: C/1996 B2 (Hyakutake) (Bockelée-Morvan et al., 1998), C/1995 O1 (Hale-Bopp) (Meier et al., 1998a) and C/2002 T7 (LINEAR) (Hutsemékers et al., 2008). The primitive Earth D/H ratio is not known. At one extreme, we have the protosolar value of the huge  $\text{H}_2$  reservoir measured from  $\text{DH}/\text{H}_2$  in Jupiter:  $(\text{D}/\text{H})_p \geq 0.26 \cdot 10^{-4}$  (Mahaffy et al., 1998). At the other end, the D/H ratio estimated to be representative of the deep mantle  $(\text{D}/\text{H})_p \leq 1.36 \cdot 10^{-4}$  (Deloule et al., 1991; Dauphas et al., 2000). In the case that water from the warm, inner solar nebula is adsorbed by the rocky grains that formed the bulk of the Earth, intermediate values  $(\text{D}/\text{H})_p \simeq 0.8\text{--}1.0 \cdot 10^{-4}$  are likely to better characterize the primitive Earth D/H ratio (Owen and Bar-Nun, 1998). With these values, the proportion of Earth's water due to comets varies between 50% and 10% (Eq. 2), with plausible values up to 30% (Eberhardt et al., 1995; Bockelée-Morvan et al., 1998; Owen and Bar-Nun, 1998). More detailed modelling, combining the D/H ratios with lunar cratering records and terrestrial mantle siderophiles, suggests that the contribution of comets to Earth's water is smaller than 10% (Dauphas et al., 2000). Moreover, should a significant fraction of cometary material possess an even higher D/H as measured in HCN (Meier et al., 1998b) and assuming that this material can be recycled at the Earth's surface, even smaller cometary contributions would be expected.

### 3 New constraints from $^{15}\text{N}/^{14}\text{N}$

Assuming similarly that the nitrogen isotopic ratio of the present Earth results from the combination of primitive Earth and cometary nitrogen mixtures, we have

$$\begin{aligned} n_p(^{14}\text{N}) + n_c(^{14}\text{N}) &= n_t(^{14}\text{N}) \\ n_p(^{15}\text{N}) + n_c(^{15}\text{N}) &= n_t(^{15}\text{N}) . \end{aligned} \quad (3)$$

and

$$\frac{n_c(^{14}\text{N})}{n_t(^{14}\text{N})} = \frac{(^{15}\text{N}/^{14}\text{N})_p - (^{15}\text{N}/^{14}\text{N})_t}{(^{15}\text{N}/^{14}\text{N})_p - (^{15}\text{N}/^{14}\text{N})_c} . \quad (4)$$

Since nitrogen is mostly in the form  $^{14}\text{N}$ , we may write

$$\frac{n_c(\text{H}_2\text{O})}{n_t(\text{H}_2\text{O})} = \frac{n_c(^{14}\text{N})}{n_t(^{14}\text{N})} \times \frac{n_t(\text{N})}{n_t(\text{H}_2\text{O})} \times \left( \frac{n_c(\text{N})}{n_c(\text{H}_2\text{O})} \right)^{-1} , \quad (5)$$

with the constraint

$$\frac{n_c(\text{H}_2\text{O})}{n_t(\text{H}_2\text{O})} \leq \frac{n_t(\text{N})}{n_t(\text{H}_2\text{O})} \times \left( \frac{n_c(\text{N})}{n_c(\text{H}_2\text{O})} \right)^{-1} \quad (6)$$

which comes from the fact that the total amount of nitrogen delivered by comets to Earth cannot exceed the amount of nitrogen presently measured in Earth's atmosphere and surface, i.e.  $n_c(^{14}\text{N})/n_t(^{14}\text{N}) \leq 1$ . Nitrogen recycling at Earth's surface is implicitly assumed as well as the fact that the composition of the atmosphere has not significantly changed since the late veneer (Tolstikhin and Marty, 1998).

The amount of water and nitrogen at the surface of the present Earth (mostly in the oceans and in the atmosphere, respectively) is relatively well known. The water inventory by Lécuyer et al. (1998) (see also Dauphas et al., 2000) gives  $1.7 \cdot 10^{21}$  kg of water at the Earth's surface (oceans, ice sheets, organic matter, metamorphic rocks, shales, sandstones, continental carbonates, evaporites, marine clays and marine carbonates). The nitrogen inventory at the Earth's surface (atmosphere, sedimentary rocks, crustal igneous rocks) gives  $5.0 \cdot 10^{18}$  kg of  $\text{N}_2$  (Zhang and Zindler, 1993). Thus  $n_t(\text{N})/n_t(\text{H}_2\text{O}) \simeq 3.8 \cdot 10^{-3}$ .

The abundance of nitrogen in comets is poorly known. Because  $\text{N}_2$  is not detected, the nitrogen inventory in cometary ices mainly relies on the mea-

surement of the  $\text{NH}_3$  and  $\text{HCN}$  volatiles. From the abundances given by Bockelée-Morvan et al. (2004), we compute  $n_c(\text{N})/n_c(\text{H}_2\text{O}) \gtrsim 10^{-2}$  adding the contributions of all observed N-bearing molecules and considering that contributions from other molecules are possible. Based on in-situ measurements by the Giotto spacecraft, Encrenaz et al. (1991) estimated the total N/O abundance in 1P/Halley:  $\text{N/O} = 0.027 \pm 0.009$ , including both the gas and the dust components. Assuming that at most 60% of O is in  $\text{H}_2\text{O}$  (Encrenaz et al., 1991),  $n_c(\text{N})/n_c(\text{H}_2\text{O}) \gtrsim 3 \cdot 10^{-2}$ . Further modelling by Greenberg (1998) and Huebner (2002) suggests that  $n_c(\text{N})/n_c(\text{H}_2\text{O})$  may be as high as  $7 \cdot 10^{-2}$ , nitrogen being still depleted in comets by a factor 3 with respect to solar abundances. Under the hypothesis that the nitrogen abundance in 1P/Halley is representative of comets, we adopt the lower limit  $n_c(\text{N})/n_c(\text{H}_2\text{O}) \gtrsim 3 \cdot 10^{-2}$  from which we derive  $n_c(\text{H}_2\text{O})/n_t(\text{H}_2\text{O}) \lesssim 13\%$  (Eq. 6).

The present Earth  $^{15}\text{N}/^{14}\text{N}$  ratio is accurately measured from  $\text{N}_2$  in the atmosphere:  $(^{15}\text{N}/^{14}\text{N})_t = 3.676 \cdot 10^{-3}$  (Junk and Svec, 1958). This is comparable to  $^{15}\text{N}/^{14}\text{N} = 3.64 \cdot 10^{-3}$  which characterizes the present mantle (Becker et al., 2003). The primitive Earth  $^{15}\text{N}/^{14}\text{N}$  is not known. It may be close to  $^{15}\text{N}/^{14}\text{N} \simeq 3.55 \cdot 10^{-3}$ , the value measured in enstatite chondrites thought to have released significant amounts of nitrogen during and shortly after the accretion phase (Javoy et al., 1986). On the other hand, a massive atmosphere could have been captured directly from the solar nebula (Pepin, 2006; Genda and Ikoma, 2008) where  $^{15}\text{N}/^{14}\text{N} \simeq 2.3 \cdot 10^{-3}$  (Owen et al., 2001; Meibom et al., 2007, this value is estimated from the atmosphere of Jupiter assumed to be a proxy of the  $\text{N}_2$  reservoir in the solar system.). A large part of the early atmosphere might have been lost during the post-accretion phase (much before the late veneer), possibly resulting in a small shift of the primitive  $^{15}\text{N}/^{14}\text{N}$  ratio due to atmospheric escape (Tolstikhin and Marty, 1998). Given these uncertainties we consider in the following both  $(^{15}\text{N}/^{14}\text{N})_p = 2.3 \cdot 10^{-3}$  and  $(^{15}\text{N}/^{14}\text{N})_p = 3.55 \cdot 10^{-3}$ . Intermediate values are possible, as well as values closer to the present terrestrial value if fractionation by atmospheric escape has been significant.

$^{15}\text{N}/^{14}\text{N}$  has been measured remotely in about twenty comets using optical-UV spectroscopy of  $\text{CN}$ , and in three comets on the basis of radio observations of  $\text{HCN}$ . Recent studies show that optical and radio measurements do agree within uncertainties (Bockelée-Morvan et al., 2008). The nitrogen isotopic ratio is remarkably similar from comet to comet and clusters around the “anomalous”  $^{15}\text{N}/^{14}\text{N} = 6.8 \pm 0.3 \cdot 10^{-3}$  which is twice as high as the terrestrial value (Manfroid et al., 2009). On the other hand, the analysis of comet 81P/Wild2 grains returned by Stardust suggests bulk  $^{15}\text{N}/^{14}\text{N}$  ratios comparable to the terrestrial and chondritic values (McKeegan et al., 2006). We therefore compute an average cometary ratio according to

$$(^{15}\text{N}/^{14}\text{N})_c = \frac{n_{ca}(\text{N})}{n_c(\text{N})} (^{15}\text{N}/^{14}\text{N})_{ca} + \left(1 - \frac{n_{ca}(\text{N})}{n_c(\text{N})}\right) (^{15}\text{N}/^{14}\text{N})_{cd} \quad (7)$$

where  $(^{15}\text{N}/^{14}\text{N})_{ca} = 6.8 \cdot 10^{-3}$ ,  $(^{15}\text{N}/^{14}\text{N})_{cd} = 3.55 \cdot 10^{-3}$ , and  $n_{ca}(\text{N})$  is the abundance of the isotopically anomalous nitrogen.

In Fig. 1 we illustrate the proportion of terrestrial nitrogen delivered by comets,  $n_c(\text{N})/n_t(\text{N})$ , as a function of the relative abundance of anomalous nitrogen  $n_{ca}(\text{N})/n_c(\text{H}_2\text{O})$  for various cases of interest (Eqs. 4 and 7). Fig. 1 shows that the nitrogen isotopic shift between primitive and present Earth can be easily accounted for by the addition of a cometary component thanks to the presence, even in small quantities, of isotopically anomalous nitrogen in comets. The proportion of terrestrial water delivered by comets can be computed from Fig. 1 and Eq. 5, i.e.  $n_c(\text{H}_2\text{O})/n_t(\text{H}_2\text{O}) \lesssim 0.13 \times n_c(\text{N})/n_t(\text{N})$  for  $n_c(\text{N})/n_c(\text{H}_2\text{O}) \gtrsim 3 \cdot 10^{-2}$ .

Assuming that the carrier of the isotopically anomalous nitrogen is only HCN, we have  $n_{ca}(\text{N})/n_c(\text{H}_2\text{O}) \simeq 2 \cdot 10^{-3}$  using the abundances of Bockelée-Morvan et al. (2004). Although  $^{15}\text{N}/^{14}\text{N}$  has not yet been measured in cometary ammonia, fractionation mechanisms predict that  $^{15}\text{N}$  must be enhanced in  $\text{NH}_3$  before being transferred to HCN compounds (Charnley and Rodgers, 2002; Rodgers and Charnley, 2008). If  $\text{NH}_3$  is also included in the carriers of the anomalous  $^{15}\text{N}/^{14}\text{N}$  ratio, we have  $n_{ca}(\text{N})/n_c(\text{H}_2\text{O}) \simeq 8 \cdot 10^{-3}$  using  $6 \cdot 10^{-3}$  for the mean abundance of  $\text{NH}_3$  relative to  $\text{H}_2\text{O}$  (Bockelée-Morvan et al., 2004). In this case between 15% and 65% of Earth's nitrogen might have been delivered by comets (Fig. 1). We then estimate the upper limit  $n_c(\text{H}_2\text{O})/n_t(\text{H}_2\text{O}) \lesssim 9\%$ , which can be as low as  $n_c(\text{H}_2\text{O})/n_t(\text{H}_2\text{O}) \lesssim 2\%$  if the primitive Earth  $^{15}\text{N}/^{14}\text{N}$  was close to  $3.55 \cdot 10^{-3}$ . Given that the abundance of nitrogen in comets is probably larger than  $n_c(\text{N})/n_c(\text{H}_2\text{O}) = 3 \cdot 10^{-2}$ , and that the refractory component may also contain isotopically anomalous nitrogen, the contribution of comets to Earth's water is probably roughly twice as small as the upper limit we just derived, i.e. not larger than a few percent.

These constraints are more stringent than the values derived from D/H using Eq. 2. Moreover, as soon as  $n_c(\text{H}_2\text{O})/n_t(\text{H}_2\text{O}) \lesssim 7\%$ , the amount of water delivered by comets is not sufficient to explain the D/H isotopic shift in terrestrial oceans from the primitive value (Eq. 2) so that additional sources of water are needed. Our estimates agree with the constraints derived from recent dynamical models of the solar system (Morbidelli et al., 2000), and from mass-balance models based on D/H (Dauphas et al., 2000) or based on noble metals and gases (Dauphas and Marty, 2002).

As far as the atmosphere is concerned, Marty and Meibom (2007) showed that a cometary contribution fitting the abundances of noble gases in the Earth's atmosphere would deliver  $\sim 6\%$  of the atmospheric nitrogen. Given the uncertainties on the abundances of noble gases in comets (Bockelée-Morvan et al.,



2004), this is compatible with our estimates provided that  $n_{ca}(\text{N})/n_c(\text{H}_2\text{O}) \simeq 10^{-2}$  and  $(^{15}\text{N}/^{14}\text{N})_p \simeq 3.55 \cdot 10^{-3}$  in Fig. 1. Early fractionation due to atmospheric escape (Tolstikhin and Marty, 1998) might also have slightly increased  $(^{15}\text{N}/^{14}\text{N})_p$  providing an even better agreement.

It is important to note that the anomalous  $^{15}\text{N}/^{14}\text{N}$  isotopic ratio was measured to be identical in Oort-cloud and Kuiper-belt comets (Hutsemékers et al., 2005; Manfroid et al., 2009) so that our estimates apply to the contributions of both types of comets.

## 4 Conclusions

Thanks to the fact that the  $^{15}\text{N}/^{14}\text{N}$  ratio measured in cometary CN and HCN is significantly different from the ratio measured on Earth, and since any delivery of nitrogen from comets to Earth necessarily accompanied that of water, we put independent constraints on the amount of Earth’s water possibly due to comets. Under reasonable assumptions, we find that no more than a few percent of Earth’s water can be attributed to comets. This is consistent with the constraints derived from D/H using various models (Morbidelli et al., 2000; Dauphas et al., 2000; Dauphas and Marty, 2002). Since the  $^{15}\text{N}/^{14}\text{N}$  isotope ratio is not different in Oort-cloud and Kuiper-belt comets, our estimates apply to both types of objects. Our results also suggest that a significant part of Earth’s nitrogen might come from comets, supporting the idea of a dual origin of the Earth’s atmosphere (Owen and Bar-Nun, 1998; Dauphas, 2003; Marty and Meibom, 2007). A critical measurement to further constrain these quantities would be the determination of the  $^{15}\text{N}/^{14}\text{N}$  ratio in cometary  $\text{NH}_3$ , expected to be either anomalous or terrestrial. Although more detailed modelling is required for more quantitative estimates, our results demonstrate the interest of considering the  $^{15}\text{N}/^{14}\text{N}$  ratio to evaluate the contribution of comets to the late bombardment of the Earth.

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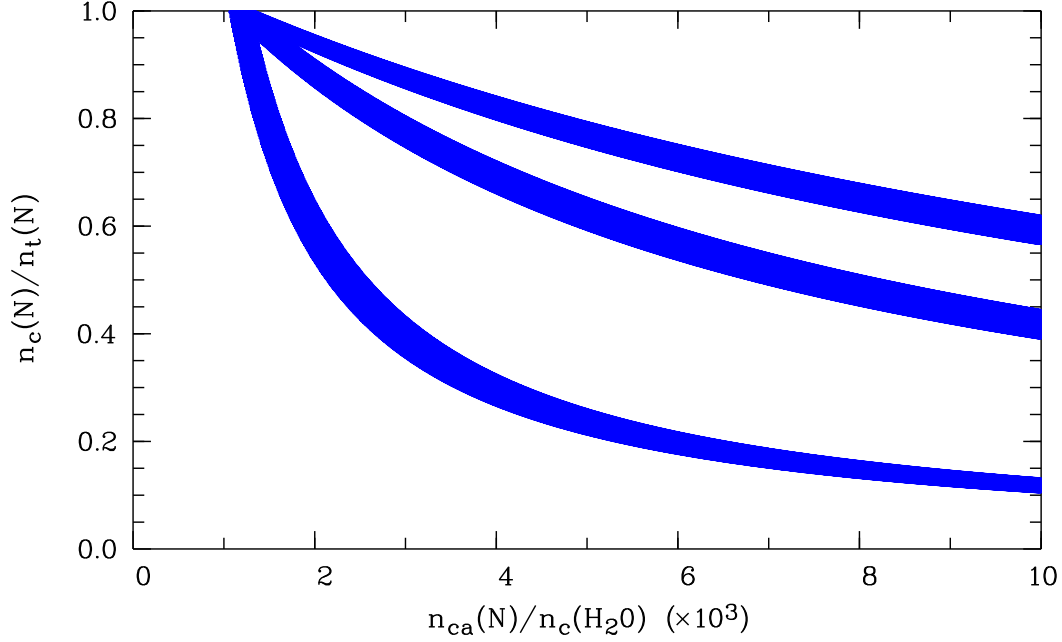


Fig. 1. The proportion of terrestrial nitrogen due to comets  $n_c(\text{N})/n_t(\text{N})$  as a function of the abundance of isotopically anomalous nitrogen in comets  $n_{ca}(\text{N})/n_c(\text{H}_2\text{O})$ . The three curves are computed from Eqs. 4 and 7 with  $n_c(\text{N})/n_c(\text{H}_2\text{O}) = 3 \cdot 10^{-2}$ . The proportion of terrestrial water due to comets is given by  $n_c(\text{H}_2\text{O})/n_t(\text{H}_2\text{O}) \lesssim 0.13 \times n_c(\text{N})/n_t(\text{N})$  (Eq. 5). Each curve corresponds to a different value of the primitive Earth nitrogen isotopic ratio :  $(^{15}\text{N}/^{14}\text{N})_p = 2.3 \cdot 10^{-3}$ ,  $3.0 \cdot 10^{-3}$  and  $3.55 \cdot 10^{-3}$  from top to bottom. The width of the curves accounts for the uncertainty on the measured cometary  $(^{15}\text{N}/^{14}\text{N})_{ca}$ .